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(54) Title: CONDUCTIVE THERMOSET MOLDING COMPOSITION AND METHOD FOR PRODUCING SAME			
(57) Abstract <p>A high filler conductive thermoset composite that may be used in bipolar plates is provided in which the composite may be used to form thin wall, electrically and thermally conductive structures that have sufficient toughness and are resistant to the harsh fuel cell environment.</p>			

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CONDUCTIVE THERMOSET MOLDING COMPOSITION AND
METHOD FOR PRODUCING SAME

This application is a continuation-in-part application of provisional application U.S. Serial No. 60/021,920, filed October 14, 1997, the entire contents of
5 which are hereby incorporated herein by reference.

BACKGROUND OF THE INVENTION

This invention relates generally to the field of thermoset composites,
10 and more particularly to methods for making structures from thermoset composites having high thermal and electrical conductivity. In addition, this invention relates to the starting materials from which such structures are formed, as well as to the methods of making such starting materials.

15 Structures having good thermal and electrical conductivity properties in a thin wall, sufficiently crack-resistant fabrication are useful for applications requiring such conductivity properties, such as for bi-polar plates for fuel cells.

A major problem long of concern to the art is ion leaching from bipolar
20 plates into the fuel cell environment. This ion leaching is problematic because it renders ineffective the membrane that separates stacked bi-polar plates in fuel cells. Ion leaching is promoted by the current charged, corrosive, acidic environment of the fuel cells. Because of ion leaching, known structures that comprise highly electrically and thermally conductive metals have largely proven ineffective for use as bi-polar
25 plates in fuel cell applications. Even known structures that incorporate substantial amounts of ionizable impurities (i.e., ionizable, conductive metals) are subject to ion leaching when used as bi-polar plates in fuel cells.

An additional problem associated with the manufacture of such structures is high cost. In an effort to solve ion leaching problem while retaining thermal and electrical conductivity properties of the resulting structure, some conventional structures have utilized exotic, high cost materials, such as carbon-
5 carbon composites, or other exotic materials. However, these exotic materials have long manufacturing times and higher raw material and labor processing costs, which are generally passed on to consumers who purchase fuel cells.

A further problem associated with such structures is their reduced
10 processibility. For example, carbon-carbon based bipolar plates are generally machined into a bipolar plate, and then handled in a time and labor-intensive manner during the fuel cell assembly process to ensure that the brittle, fragile carbon-carbon thin bipolar plate does not crack or break.

15 Another problem associated with such structures is the potential cracking associated with thin wall structures that may not have sufficient toughness to resist cracking. For example, carbon-carbon based bipolar plates are generally relatively brittle, and may be subject to cracking and/or breaking in the fuel cell environment.

20

An additional problem associated with such structures is the reduction in thermal and electrical conductivity found in structures unable to incorporate sufficient conductive materials. For example, known molding compositions incorporating conductive materials are generally characterized by an electrical
25 resistivity (i.e., the reciprocal of electrical conductivity) of not less than 0.005 Ohm-cm.

Accordingly, there is a need for new and better structures having good thermal and electrical conductivity properties in a thin wall, sufficiently crack-

resistant fabrication that exhibit low ion leaching, low manufacturing costs, and increased processibility. Additionally, there is a need for new and better methods of making such structures at lower manufacturing and processing costs. Further, there is a need for new and better starting materials for such structures, and new and better
5 methods of making such starting materials.

SUMMARY OF THE INVENTION

In accordance with the invention, it has been discovered that thermosetting resin systems may be sufficiently packed with low cost fillers to make a molding composition that can be formed into structures exhibiting good electrical and
10 thermal conductivity, low ion leaching, low manufacturing costs, increased processibility, and enhanced crack-resistance. Ways of making this molding composition to promote the electrical and thermal conductivity properties in a structure formed therefrom have also been developed.

Accordingly, under one aspect of the invention, there have been
15 developed thermosetting molding compositions comprising about 10 to about 30 wt-wt percent of a low viscosity thermosetting binder resin system, and about 70 to about 90 wt-wt percent of a filler.

In a further aspect of the invention, there have also been developed methods of making a thermosetting molding composition comprising mixing a low
20 viscosity thermosetting binder resin system and a filler under conditions selected to

preserve the integrity of the filler and under conditions sufficient to wet-out the conductive filler with the binder resin system.

In an additional aspect of the invention, there have also been developed electrically and thermally conductive structures made from the foregoing
5 thermosetting molding compositions.

In another aspect of the invention, there have been developed methods of making a structure from the foregoing thermosetting molding compositions, wherein the methods comprise molding into a desired shape the foregoing thermosetting molding compositions, and subjecting the desired shape to conditions
10 sufficient to substantially cure the foregoing thermosetting molding compositions to form a structure.

The invention has the following advantages. The invention provides structures that exhibit properties of enhanced crack-resistance in thin wall structures, increased processibility, low ion leaching, low manufacturing costs, and enhanced
15 electrical conductivity and thermal conductivity. Further, invention molding compositions are non-reactive in the corrosive fuel cell environment, thereby promoting low ion leaching. In addition, invention molding compositions comprise thermosetting resins which have sufficiently low viscosity to enhance the processibility of the molding composition at moderate temperatures and pressures,
20 thereby promoting the formation of thin wall structures for use in fuel cell applications. Also, the invention methods of making molding compositions

optionally enhance the thermal conductivity (i.e., thermal conductivity of no less than 3 (W/m) K) and electrical conductivity (i.e., electrical resistivity of no greater than 0.01 Ohm-cm) of structures formed from the molding composition, thereby promoting use of these structures (i.e., as bipolar plates) in fuel cells.

5 The invention provides a molding composition that can be used to manufacture bi-polar plates for use in a fuel cell. The molding composition may be formed into a thin-walled structure having adequate toughness to resist cracking and resistance to ion leaching to survive in the current charged, corrosive, acidic environment of a fuel cell. The molding composition also has high electrical and
10. thermal conductive properties. Because the molding composition can be manufactured with low raw material costs, short manufacturing process cycle times, and fast cure times, it can be economically used to produce bi-polar plates for lower volume, higher cost stationary fuel cells, as well as for the higher volume, lower cost fuel cells suitable to the automotive industry.

15 DETAILED DESCRIPTION OF THE INVENTION

In accordance with the invention, there are provided thermosetting molding compositions comprising a low viscosity thermosetting binder resin system, and a filler.

The low viscosity thermosetting binder resin system can be present in
20 different concentrations depending on the end use application for the invention molding composition. For example, the low viscosity thermosetting binder resin

system is preferably present within the range from about 10 to about 30 wt-wt percent of the invention composition, for example, a range of about 15 to about 25 wt-wt percent of the invention composition, and a presently preferred range of about 19 to about 23 wt-wt percent of the invention composition.

5 Similarly, the filler may be present in different concentrations depending on the end use for the invention molding composition. For example, the filler is preferably present within the range from about 70 to about 90 wt-wt percent of the invention composition, for example, a range of about 75 to about 85 wt-wt percent of the invention composition, with a presently preferred range of about 77 to about 81
10 wt-wt percent of the invention composition.

As utilized herein, "wt-wt percent" means the amount of a particular component of present in a combination, when measured as a percentage of the total weight of the combination. Thus, for example, the term "about 70 to about 90 wt-wt percent of a filler" when used to describe the amount of that component of "[a]n
15 electrically and thermally conductive thermosetting resin composition" means that the amount of the filler present in the conductive thermosetting composition may be any value in the range from about 70 percent to about 90 percent of the weight of the conductive thermosetting molding composition. Similarly, the term "the filler is ... present within [a particularly defined range of] wt-wt percent of the invention
20 composition" means that the amount of the filler present in the conductive thermosetting composition may be any value in the particularly defined range of the weight of the conductive thermosetting molding composition.

Fillers utilized in accordance with the invention include natural graphite flake, synthetic graphite, any other type of filler that, when mixed with a low viscosity thermosetting binder resin system under conditions selected to preserve the integrity of the filler to form a thermosetting molding composition, can be used to
5 produce an electrically and thermally conductive structure having a thermal conductivity in the range from 3 (Watts/meter) Kelvin [(W/m) K] to 50 (W/m) K and/or an electrical resistivity in the range from about 0.0001 Ohm cm to about 0.01 Ohm cm, and suitable combinations thereof. Preferred fillers include natural graphite flakes. Optionally, fillers may be electrically and thermally conductive.

10 Fillers may have varying shapes, size classifications, thicknesses, and purity levels.

Optionally, the fillers have an average size classification (mesh size) within the range from about 20 microns to about 4.75 mm, with a presently preferred range of about 200 microns to about 400 microns.

15 Further, the fillers optionally have an average thickness within the range from about 5 microns to about 100 microns, with a presently preferred range of about 10 microns to about 50 microns.

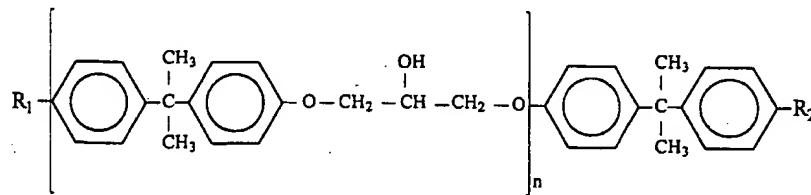
In addition, carbon-based conductive fillers (e.g., natural graphite flake, synthetic graphite and the like) optionally have an average purity (i.e.,
20 percentage carbon content, as determined on a wt-wt percent basis) within a preferred range from about 90 percent pure to about 100 percent pure, with a presently preferred

range of about about 95 percent pure to about 100 percent pure. Those carbon-based conductive fillers of lower purity result in formed structures that have proportionally lower conductivity and that are more prone to ion leaching.

Low viscosity thermosetting binder resin systems useful in the practice of the invention comprise the following precursors: epoxy vinyl ester resin systems, a low molecular weight phenolic resin system (optionally containing an amine curing agent), and combinations of a low viscosity epoxy resin and a low molecular weight phenolic resin curing agent wherein the stoichiometric ratio of the phenolic resin curing agent to the epoxy resin is optionally a predetermined phenolic-epoxy stoichiometric ratio, suitable combinations thereof, and the like.

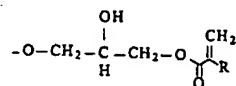
In accordance with the invention, precursors that comprise the low viscosity thermosetting binder resin systems include epoxy vinyl ester resin systems. epoxy vinyl ester resin systems contemplated for use in the practice of the invention include epoxy vinyl ester resins and the like. Epoxy vinyl ester resins include bis-A type epoxy vinyl ester resins, novolac type epoxy vinyl ester resins, suitable combinations thereof, and the like.

"Bis-A type epoxy vinyl ester resins" are resins having the following generalized structure:



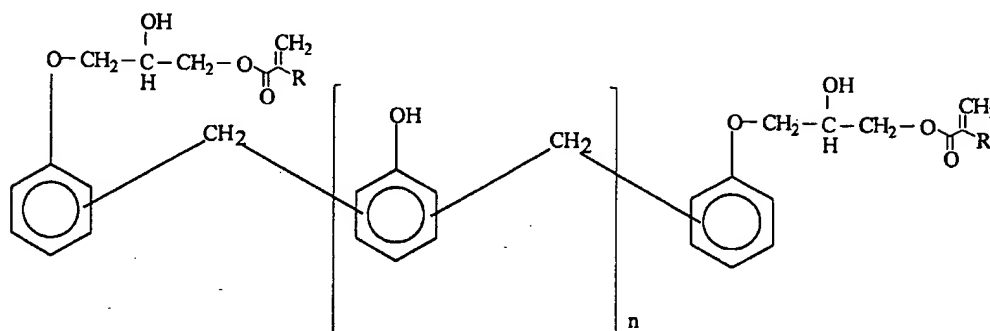
wherein the value for $n = 0-5$,

wherein R_1 and R_2 are each independently selected from:



wherein each R is independently selected from hydrogen and methyl.

- 5 “Novolac type epoxy vinyl ester resins” are resins having the following generalized structure:



wherein the average value for $n = 0-4$, and

wherein each R is independently selected from hydrogen and methyl.

- 10 Epoxy vinyl ester resins contemplated for use in the invention optionally have a viscosity within the range from about 200 centipoise to about 500 centipoise at 25°C, with a presently preferred range of about 250 centipoise to about 350 centipoise at 25°C.

- The viscosity of any of the precursors which comprise the low
- 15 viscosity thermosetting binder resin systems may be measured by any means known in the art for determining the viscosity at a given temperature. For example, the

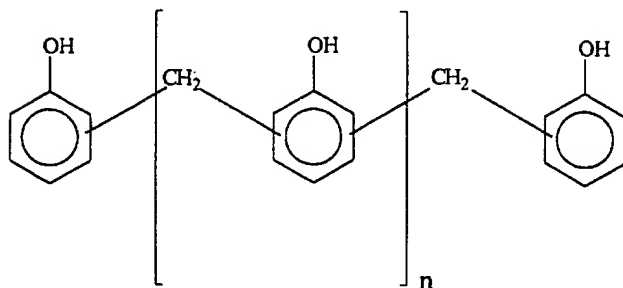
viscosity may be measured by the rotational viscometer procedure. The rotational viscometer procedure is published in the Annual Book of ASTM Standards (1997), under the fixed designation D 2196-86, and under the title "Standard Test Methods for Rheological Properties of Non-Newtonian Materials by Rotational (Brookfield) Viscometer." All portions of the Annual Book of ASTM Standards relevant to this rotational viscometer procedure are hereby incorporated herein by reference.

Exemplary epoxy vinyl ester resins include Derakane 470-300™, Derakane 411-350™, Derakane 441-400™, Derakane 510A-40™, and Derakane 510C-350™ (each of which is commercially available from Dow Chemical Corporation, Midland, MI), and the like, and suitable combinations thereof.

In accordance with the invention, precursors which comprise the low viscosity thermosetting binder resin systems include a low molecular weight phenolic resin system optionally containing an amine curing agent.

Low molecular weight phenolic resin systems utilized in the practice of the invention include phenolic novolac-based resins, resole-based resins, suitable combinations thereof, and the like.

“Phenolic novolac-based resins” are resins that have the following generalized structure:

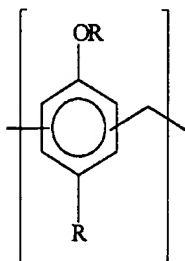


wherein the average value for $n = 0-4$

5 “Phenolic novolac-based resins” contemplated for use in the practice of the invention optionally have a softening point within the range from about 60°C to about 140°C, with a presently preferred range of about 60 °C to about 90 °C.

The softening point of any of the precursors which comprise the low viscosity thermosetting binder resin systems is a temperature at which the particular precursor softens. The softening point may be measured by any means known in the art for determining the softening point. For example, the softening point may be measured by the ring-and-ball procedure. The ring-and-ball procedure is published in the Annual Book of ASTM Standards (1997), under the fixed designation E 28-97, and under the title “Standard Test Methods for Softening Point by Ring-and-Ball Apparatus.” All portions of the Annual Book of ASTM Standards relevant to this ring-and-ball procedure are hereby incorporated herein by reference. In many instances, the softening point may be an indirect measure of the average molecular weight of the particular precursor.

“Resole-based resins” are resins which are reaction products of formaldehyde and phenol in a mole ratio of formaldehyde to phenol of no less than 100%, which reaction and reaction products are well known in the art. “Resole-based resins” include resins having the following generalized structure, and may or may not have a repeating unit present:

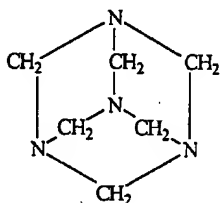


where each R is independently selected from hydrogen, formaldehyde, phenol, condensation products of formaldehyde and phenol, and suitable condensation products thereof.

“Resole-based resins” contemplated for use in the practice of the invention optionally have a viscosity (which varies as a function of the solid content) within the range from about 100 centipoise to about 40000 centipoise (which corresponds to a solids content within the range of 50% to 90%) at 25°C, with a presently preferred range of about 180 centipoise to about 300 centipoise (which corresponds to a solids content within the range of 60% to 64%) at 25°C.

Exemplary resole-based resins include SC 1008 (commercially available from Borden Chemical, Louisville, KY) and the like.

An amine curing agent may optionally be added to low viscosity resin binder systems comprising phenolic novolac-based resins. These amine curing agents include hexamethylenetetraamine and the like. Hexamethylenetetraamine has the following structure:



5

Optionally, hexamethylenetetraamine may be present in an amount within the range from about 4 and about 12 wt-wt percent of the invention composition, with a presently preferred amount of hexamethylenetetraamine of about 7 wt-wt percent of the invention composition.

10

In accordance with the invention, precursors which comprise the low viscosity thermosetting binder resin systems include combinations of low viscosity epoxy resins and low molecular weight phenolic resin curing agents where the stoichiometric ratio of the phenolic resin curing agent to the epoxy resin is optionally within the range from about 60% to about 110%, for example, a range of about 65%

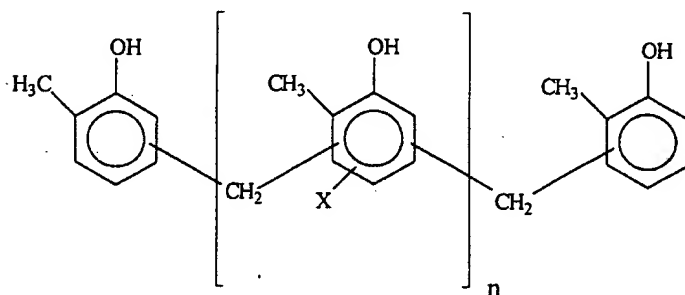
15 to about 100%, and a presently preferred range of about 65% to about 90%..

Low viscosity epoxy resins utilized in accordance with the invention include phenolic novolac-based resins, cresolic novolac-based resins, epoxidized phenolic novolac-based resins, epoxidized cresolic novolac-based resins, bi-phenyl-based epoxy resins, dicyclopentadiene-based epoxy resins, bis-F type epoxy resins,

bis-A type epoxy resins, any of the foregoing resins in solid or liquid form, and combinations thereof. Preferred low viscosity epoxy resins include epoxidized phenolic novolac-based resins, with a presently preferred low viscosity epoxy resin of DEN 431™ and DEN 438™ (commercially available from Dow Chemical Corp. 5 Midland, MI).

“Phenolic novolac-based resins” are resins having the previously defined generalized structure.

“Cresolic novolac-based resins” are resins having the following generalized structure:



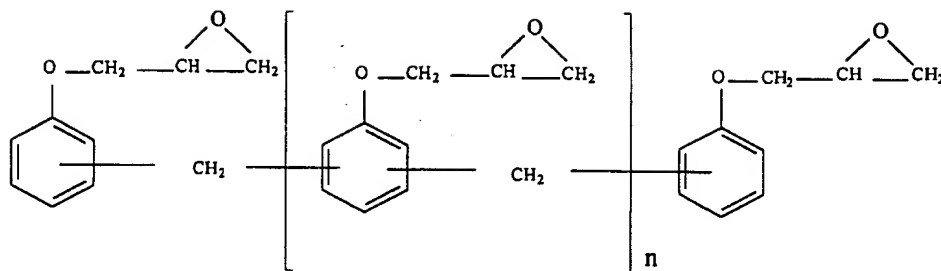
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wherein the average value for $n = 0-3$, and

wherein each X is independently selected from hydrogen and methyl.

“Epoxidized phenolic novolac-based resins” are resins having the 15 following generalized structure:

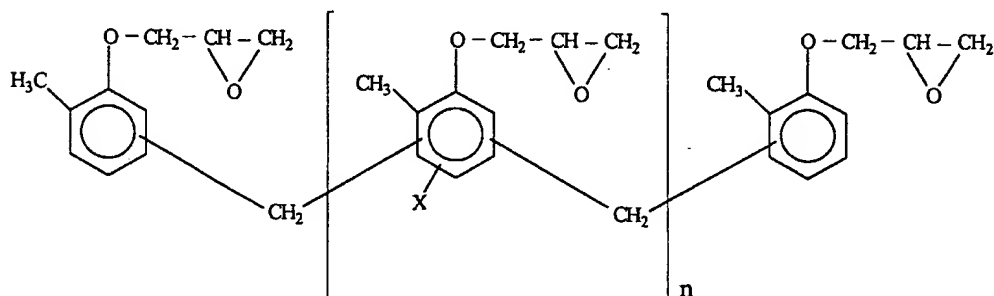
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wherein the average value for $n = 0-2$.

“Epoxidized cresolic novolac-based resins” are resins having the

5 following generalized structure:



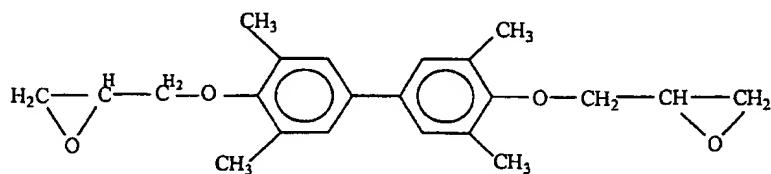
wherein the average value for $n = 0-2$, and

wherein each X is independently selected from hydrogen and methyl.

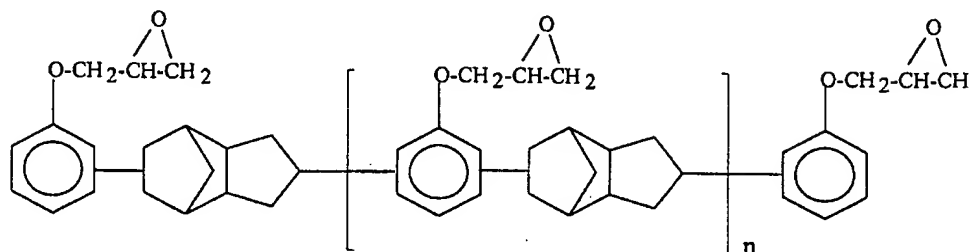
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“Bi-phenyl-based epoxy resins” are resins that have the following

structure:



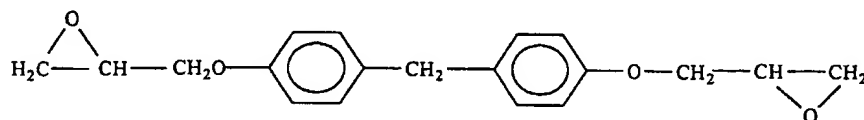
"Dicyclopentadiene-based epoxy resins" are resins that have the following structure:



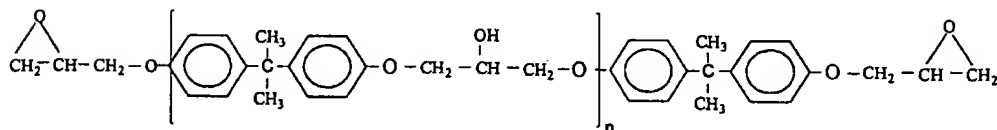
wherein the average value for $n = 0-1$.

5

"Bis-F type epoxy resins" are resins that have the following structure:



"Bis-A type epoxy resins" are resins that have the following structure:



10 wherein the average value for $n = 0-5$.

Low viscosity epoxy resins utilized in the practice of the invention optionally have a viscosity within the range from about 1400 centipoise to about 50000 centipoise at 52°C. Exemplary ranges include a viscosity range of 2000 centipoise to 15000 centipoise at 52°C, and a presently preferred viscosity range of 4000 centipoise to 12000 centipoise at 52°C.

15

In addition, the low viscosity epoxy resins contemplated for use in the practice of the invention optionally have an epoxy equivalent weight (EEW) within the range from about 165 to about 250. Preferred low viscosity epoxy resins (i.e., epoxidized phenolic novolac-based resins) have a preferred EEW range of 170 to 190, 5 and a presently preferred EEW of 172 to 181.

Further, the low viscosity epoxy resins optionally have a glass transition temperature (T_g) within the range from about -40°C to about 170°C .

Exemplary low viscosity epoxy resins utilized in accordance with the invention include DEN 431TM, DEN 438TM (each of which is commercially available 10 from Dow Chemical Corporation, Midland, MI), DER 661TM (commercially available from Dow Chemical Corporation, Midland, MI), and Epon 828TM (commercially available from Shell Development Co., Houston, TX).

Low molecular weight phenolic resin curing agents useful in the practice of the invention include phenolic novolac-based resins.

15 Low molecular weight phenolic resin curing agents contemplated for use in the practice of the invention optionally have a softening point within the range from about 60°C to about 90°C , for example a range of about 75°C to about 85°C , and a presently preferred range of about 80°C to about 85°C .

Further, the low molecular weight phenolic resin curing agents utilized 20 in accordance with the invention optionally have a molecular weight within the range

from about 200 to about 600, for example, a range of about 300 to about 500, and a presently preferred range of about 370 to about 420.

In addition, the low molecular weight phenolic resin curing agents utilized in the practice of the invention optionally have a hydroxy equivalent weight
5 (HEW) within the range from about 95 to about 115, for example, a HEW range of 100 to 110, and a presently preferred HEW range of 102 to 106.

Examples of the low molecular weight phenolic resin curing agents useful in accordance with the invention include HRJ 1166™ and HRJ 1583™, each of which is commercially available from Schenectady International, Inc. (Schenectady,
10 N.Y.).

In accordance with the invention, the stoichiometric ratio of the low molecular weight phenolic resin curing agent to the low viscosity epoxy resin is optionally within the range from about 60% to about 110%, for example, a range of about 65% to about 100%, and a presently preferred range of about 65% to about
15 90%.

As utilized herein, the term "stoichiometric ratio" means the ratio of the number of curing agent functionalities (e.g., hydroxy functionalities) to the number of resin functionalities (e.g., epoxy functionalities) available in the precursors which comprise the low viscosity thermosetting binder resin system. The number of a
20 particular functionality may be determined by means known in the art. For example, the number of epoxy functionalities in the low viscosity thermosetting binder resin

system may be determined by dividing the weight of the epoxy-based precursor component (e.g., low viscosity epoxy resin) of the low viscosity thermosetting binder resin system by the epoxy equivalent weight (EEW) of the epoxy-based precursor component thereof. "Epoxy equivalent weight" means the weight (in grams) of a precursor (e.g., low viscosity epoxy resin) that contains 1 gram equivalent of epoxy functionality. Similarly, the number of hydroxy functionalities in the low viscosity thermosetting binder resin system may be determined by dividing the weight of the hydroxy-based precursor component (e.g., low molecular weight phenolic resin; low molecular weight phenolic resin curing agents) of the low viscosity thermosetting binder resin system by the hydroxy equivalent weight (HEW) of that hydroxy-based precursor component. "Hydroxy equivalent weight" means the weight (in grams) of the precursor (e.g., low molecular weight phenolic resin curing agent) which contains 1 gram equivalent of hydroxy functionality.

The low viscosity thermosetting binder resin system useful in the practice of the invention may have further optional constituents.

For example, the low viscosity thermosetting binder resin system may include a suitable catalyst system present in a wt-wt percent range from about 0.001 wt-wt percent to about 3 wt-wt percent of the invention composition. Suitable catalyst systems may vary depending on the resin and/or curing agent constituents of the low viscosity thermosetting binder resin system.

Suitable catalyst systems for use with an epoxy vinyl ester resin system include methyl ethyl ketone peroxide, cumene hydroperoxide, dimethylaniline, and the like, and suitable combinations thereof.

Suitable catalyst systems for use with a low molecular weight phenolic resin system (e.g., a phenolic novolac-based resin) optionally containing an amine curing agent include acids (with associated pHs) such as toluene sulfonic acid, phenol sulphonic acid, and the like, and suitable combinations thereof.

For use with a combination of a low viscosity epoxy resin and a low molecular weight phenolic resin curing agent, the suitable catalyst system is selected to advance crosslinking of the low viscosity epoxy resin with the low molecular weight phenolic resin. Such catalysts include urea-based catalysts, imidazole catalysts, tri-phenyl phosphine, phosphonium salt catalysts, tertiary amine catalysts, amine salt catalysts, and the like, and suitable combinations of any of the foregoing. Exemplary urea-based catalysts include diuron, monuron, phenuron, suitable combinations thereof, and the like. Exemplary imidazole catalysts include 2-phenyl-4-methyl imidazole (2P4MZ), 2-methyl imidazole, 2-phenyl imidazole, suitable combinations thereof, and the like. Exemplary tertiary amine catalysts include benzyl dimethylamine (BDMA) and the like. Exemplary amine salt catalysts include benzyl trimethyl ammonium chloride and the like.

Mixtures of catalysts may also be utilized. Thus, a preferred suitable catalyst system for the low viscosity thermosetting binder resin system comprising a

combination of a low viscosity epoxy resin and a low molecular weight phenolic resin curing agent comprises a urea-based catalyst (e.g., diuron) present in a wt-wt percent range from about 0.001 wt-wt percent to about 1.5 wt-wt percent of the invention composition, and an imidazole catalyst (e.g., 2-phenyl - 4 - methyl imidazole) present
5 in a wt-wt percent range from about 0.001 wt-wt percent to about 0.5 wt-wt percent of the invention composition. Exemplary mixtures include a presently preferred mixture of about 0.5 to about 0.7 wt-wt percent of diuron and about 0.03 to about 0.1 wt-wt percent of 2-phenyl - 4 - methyl imidazole.

When utilized in the practice of the invention, suitable catalyst systems
10 permit the conductive thermosetting molding composition to substantially cure in no more than about 90 seconds, for example, no more than about 60 seconds, and preferably no more than about 45 seconds, at suitable curing conditions.

As utilized herein, the term "substantially cure" means that the curing of the molding composition has advanced sufficiently so that at least 50 percent of the
15 available functionalities have been reacted from the binder resin component having the lower stoichiometric availability.

The percentage level of cure of a material may be measured by procedures known in the art. For example, differential scanning calorimetry (DSC) may be used to measure levels of cure. Under DSC, uncured material and material to
20 be tested for substantial curing are subjected to increasing temperatures over time, and the heat output from the material is measured as a function of this increasing

temperature. Since epoxy resin - phenolic resin polymerization is an exothermic reaction, relevant areas of the DSC curve (heat flow as a function of temperature) measure the amount of uncured material from the resin component having the lower stoichiometric availability (i.e., the residual cure level). Thus, when the relevant area
5 of the DSC curve for material being tested for substantial curing (i.e. the residual cure level) is compared to the relevant area of the DSC curve for an uncured material (i.e., the total cure level), the ratio of the residual cure level to the total cure level, when normalized for tested weights of material, provides the percentage of available functionalities that have not been reacted from the resin component having the lower
10 stoichiometric availability in the material to be tested. This percentage subtracted from 100 percent provides the percentage of available functionalities that have been reacted to determine whether the material to be tested has substantially cured.

Suitable curing conditions include conditions that advance the formation of derivatives from the precursors of the low viscosity binder resin system
15 (i.e., curing), such as temperature, pressure, other reaction conditions (e.g., presence of high energy photons, and the like), and suitable combinations thereof. At suitable curing conditions, the precursors of the low viscosity thermosetting binder resin systems form derivatives having the general chemical formula (precursor)_m minus the particular leaving groups required for the formation of the applicable derivative of the
20 precursor, wherein $m > 1$. These derivatives include any of the following applicable reaction products of the precursors: oligomers, glycidyl amines, ethoxylated species, cross-linked species (e.g., addition products, including etherification, and

condensation products), and chain-extended species (e.g., addition products, including etherification, and condensation products), and the like. Those of skill in the art can readily determine which reaction products are applicable to a particular precursor.

Exemplary means for providing such suitable curing conditions include

5 injection molding, transfer molding, and compression molding, each under suitable molding temperatures and pressures. Suitable molding temperatures include temperatures in a range from about 250°F to about 450°F, for example, a range of about 300°F to about 375°F, and a presently preferred range of about 325°F to about 350°F. Suitable molding pressures include pressures in a range from about 100 psi

10 (pounds per square inch) to about 3000 psi, for example, a range of about 400 psi to about 1500 psi, and a presently preferred range of about 500 psi to about 1000 psi.

In addition, the low viscosity thermosetting binder resin system may further optionally comprise an internal release agent present in a wt-wt percent range from about 0.001 wt-wt percent to about 2 wt-wt percent of the invention

15 composition. As utilized herein, the term "internal release agent" means any composition which keeps the thermosetting molding composition from adhering to the surface of a mold in which the thermosetting molding composition may be shaped into a desired shape to form a structure. Exemplary internal release agents include Carnuba wax, zinc stearate, calcium stearate, fatty acids (and derivatives thereof),

20 suitable combinations thereof, and the like.

Further, the low viscosity thermosetting binder resin system may optionally comprise a binder resin system solvent present in a wt-wt percent range from about 0.001 wt-wt percent to about 15 wt-wt percent of the invention composition, for example, in a range from about 7 wt-wt percent to about 11 wt-wt percent of the invention composition. As utilized herein, the term "binder resin system solvent" includes any solvent in which the low viscosity thermosetting binder resin system may be at least partly soluble or miscible, and serves to further decrease the viscosity of the thermosetting molding composition. Exemplary binder resin system solvents include acetone, methyl ethyl ketone, dichloromethane, suitable combinations thereof, and the like.

In accordance with the invention, there are further provided methods of making a thermosetting molding composition comprising mixing a binder resin system and a conductive filler under conditions selected to preserve the integrity of the filler and under conditions sufficient to wet-out the conductive filler with the binder resin system. Optionally, the thermosetting molding composition formed thereby may be heated to a temperature in the range from the boiling point of the binder resin system solvent to less than about the cure temperature of the binder resin system, for a sufficient time to reduce the level of binder resin system solvent to less than 3 wt-wt percent of the conductive thermosetting composition.

Conditions selected to preserve the integrity of the filler include conditions wherein the shear force to which the binder resin system and conductive filler are subjected (i.e., during mixing of these two components of the invention

composition) are sufficiently low so that a structure made from the resulting molding composition has an electrical resistivity of no more than 0.005 Ohm-cm.

It has been discovered that too much work applied in mixing the binder resin system and conductive filler (i.e., during mixing of these two components of the invention composition) reduces the integrity of the filler particles, resulting in a structure made from the resulting molding composition that has an electrical resistivity of no less than about 0.005 Ohm-cm. Accordingly, conditions selected to preserve the integrity of the filler also include conditions where the work applied in mixing the binder resin system and conductive filler (i.e., during mixing of these two components of the invention composition) is sufficiently low that a structure made from the resulting molding composition has an electrical resistivity of no more than 0.005 Ohm-cm. This work may be determined by measuring (i.e., by methods known in the art) the electrical energy input over time (normalized to batch size) into a mixer used in the mixing process under particular reaction conditions. As contemplated in the invention, the work required to preserve the integrity of the filler would not exceed the work generated by a Hobart Mixer Model No. V-1401 (commercially available from Hobart, Inc., Troy, OH), operating at about 70°F at speed setting number 3 (i.e., about 150 rpm) for about 30 minutes and utilizing a blade configuration T68064 (flat beater) (commercially available from Hobart, Inc., Troy, OH), in mixing the batch size and components of the thermosetting molding composition of Example 6.

In addition, exemplary conditions selected to preserve the integrity of the filler include a spraying of the binder resin system on to the conductive filler as part of a continuous processing mechanism (e.g., continuous conveyor belt), a folding (e.g., in a tumbler) of the conductive filler into the binder resin system, and the like.

5 As utilized herein, the term "wet-out" means to intimately mix the binder resin system with or fully disperse the binder resin system within the filler so that there are no observable (to the naked eye) uncoated conductive filler flakes. At the time of wet-out, the conductive filler flakes may become sticky (due to the binder resin coating), and these conductive filler flakes may no longer form a pourable
10 solution. Exemplary conditions that promote "wet-out" include increased mixing time, increased mixing speed, addition of the optional binder system solvent to the mixture of the binder resin system and the conductive filler, and increased mixing and processing temperature to a temperature below the cure temperature of the binder resin system.

15 In an additional aspect of the invention, there are also provided electrically and thermally conductive structures made from the foregoing thermosetting molding compositions. Optionally, such a structure may comprise at least one thin wall section in any desired polygonic or curvilinear shape. A preferred shape of the thin wall section is any shape which promotes use of the structure, which
20 comprises the thin wall section, as a bipolar plate.

The thickness of the thin wall section may be in the range from about 0.003 inches to about 0.200 inches, with a preferred range of about 0.012 inches to about 0.060 inches. This thickness may be measured by methods known in the art (i.e., micrometer, calipers, and the like).

5 The thin wall section of the foregoing structure may further possess additional optional properties. For example, the thin wall section of the structure optionally has strength sufficient to avoid cracking (i.e., visible cracks and/or microfractures) when under a flexural stress of no less than about 50% of the structure's ultimate flexural strength. Optionally, the thin wall section of the structure
10 has strength sufficient to avoid cracking (i.e., visible cracks and/or microfractures) when under a stress of no more than about 95% of the structure's ultimate flexural strength. An exemplary range for the flexural stress is in a range from about 2500 psi to about 7200 psi.

 The ultimate flexural strength of a structure may be measured by any
15 means known in the art for determining flexural strength. For example, the ultimate flexural strength may be measured by an ASTM flexural property procedure. The ASTM flexural property procedure is published in the Annual Book of ASTM Standards (1997), under the fixed designation D790-97, and under the title "Standard Test Methods for Flexural Properties of Unreinforced and Reinforced Plastics and
20 Electrical Insulating Materials." All portions of the Annual Book of ASTM Standards relevant to this ASTM flexural property procedure are hereby incorporated herein by reference.

In addition, the thin wall section of the structure optionally has an electrical resistivity of no more than about 0.0025 Ohm-cm. Optionally, the thin wall section of the structure has an electrical resistivity of no less than about 0.0001 Ohm-cm. Exemplary ranges include a range from about 0.0002 Ohm-cm to about 0.0025 Ohm-cm, with a presently preferred range from about 0.00035 Ohm-cm to about 0.001 Ohm-cm.

The electrical resistivity may be measured by any means known in the art for determining electrical resistivity. For example, the electrical resistivity of a structure may be measured by an ASTM electrical resistivity measurement procedure.

10 The ASTM electrical resistivity measurement procedure is published in the Annual Book of ASTM Standards (1997), under the fixed designation B193-95, and under the title "Standard Test Methods for Resistivity of Electrical Conductor Materials." All portions of the Annual Book of ASTM Standards relevant to this ASTM electrical resistivity measurement procedure are hereby incorporated herein by reference.

15 Further, the thin wall section of the structure may have a thermal conductivity (as tested as an average over a temperature range from about 20°C to about 120°C) of no less than about 3 (W/m) K. Optionally, the thin wall section of the structure has an electrical resistivity of no less than about about 50 (W/m) K. Exemplary ranges include a range from about 4.5 (W/m) K to about 20 (W/m) K, with

20 a presently preferred value of about 6 (W/m) K to about 15 (W/m) K.

The thermal conductivity may be measured by any means known in the art for determining thermal conductivity. For example, the thermal conductivity of a structure may be measured by the flash method. The flash method is published in the Annual Book of ASTM Standards (1997), under the fixed designation E1461-92, and
5 under the title "Standard Test Method for Thermal Diffusivity of Solids by the Flash Method." All portions of the Annual Book of ASTM Standards relevant to this flash method are hereby incorporated herein by reference.

In a further aspect of the invention, there are provided methods of making a structure from the foregoing thermosetting molding compositions, the
10 method comprising molding into a desired shape the foregoing thermosetting molding composition, and subjecting the desired shape to suitable curing conditions to form a structure.

Molding of the structures is accomplished by any method that does not increase the electrical resistivity of a thin wall structure formed from the invention
15 molding composition to a value greater than about 0.005 Ohm-cm, including injection molding, transfer molding, compression molding, and any other method of molding that can be performed under conditions selected to preserve the integrity of the filler.

Optionally, the desired shape can be substantially cured by the foregoing method of making a structure in no more than about 60 seconds at suitable
20 curing conditions. Optionally, the molding composition which has been substantially cured may be further cured by continued treatment at suitable curing conditions (e.g.,

baked in oven at about 350°F) for a sufficient time to substantially completely polymerize the binder resin component of the invention molding composition (e.g., for about 1 to about 10 minutes).

All references cited herein are hereby incorporated herein by reference.

- 5 The invention will now be described in greater detail with reference to the following non-limiting examples. Those of ordinary skill in the art, when guided by the teachings of the specification, may discover during the term of this patent other embodiments of the invention which fall within the scope of the appended claims.

Example 1Method of Making Thermosetting Molding Composition

5

The thermosetting molding composition identified in Examples 2 - 5, and in Example 6 (subject to the modifications noted in Example 6), was made as follows.

A specified amount of low viscosity epoxy resin (e.g., DEN 431™, 10 DEN 438™, or mixtures thereof) was added to the mixing bowl of a Hobart Mixer, Model No. A200 (commercially available from Hobart, Inc., Troy, OH). The mixer was set to operate at speed setting number 3 during the entire mixing process required to make the electrically and thermally conductive thermosetting molding composition.

A specified amount of the low molecular weight phenolic resin (e.g., 15 HRJ-1166, HRJ-1583, or mixtures thereof) was dissolved in a specified amount of the binder resin system solvent. This phenolic resin/solvent solution was slowly added to the low viscosity epoxy resin in the mixer.

A first internal release agent (e.g., Carnuba wax), a urea-based catalyst (e.g., diuron), a second internal release agent (e.g., zinc stearate), and an imidazole 20 catalyst (e.g., 2-phenyl-4-methyl imidazole) were each slowly added to the mixture in the mixer one at a time.

The resultant mixture, which provides a completed low viscosity thermosetting binder resin system, was removed from the mixer when completely mixed, and set aside.

A specified amount of conductive filler (e.g., natural graphite flake) was added to the mixing bowl of the mixer.

A specified amount of the low viscosity thermosetting binder resin system prepared, removed from the mixer, and set aside as described above was
5 slowly added to the conductive filler in the mixer. The resulting thermosetting molding composition was mixed in the mixer until the conductive filler was wet-out by the binder resin system (e.g., 1-3 minutes for 2500 gram batch size).

The resulting wet thermosetting molding composition was tray dried (e.g., oven dried) at 180°F until such time as the binder solvent level (e.g., acetone)
10 was below 3 wt-wt percent of the molding composition, as measured by residual volatile loss of a sample of the molding composition when heated to 325°F for 20 minutes.

Example 2DEN 431™-Based Thermosetting molding Composition

- 5 A 2500 gram batch of DEN 431™-based thermosetting molding composition was prepared according to the method of Example 1 so as to comprise the components shown in Table 1 below:

Table 1

10

Component	% by Weight (Normalized for Acetone)
DEN 431™ Liquid Epoxy Resin	13.08 (± 2%)
HRJ 1166 Phenolic Novolac	3.41 (± 1 %)
Carnuba Wax	0.51 (± 0.1 %)
Diuron	0.51 (± 0.01 %)
Zinc Stearate	0.34 (± 0.1 %)
Acetone (Carrier/diluent solvent)	8.68 (± 0.5 %)
Natural Flake Graphite	73.06 (-5% to +15 %)

Example 3DEN 431TM/438TM/Lower Imidazole-Based
Thermosetting Molding Composition.

- 5 A 2500 gram batch of DEN 431TM/438TM/Imidazole (2P4MZ) (0.044 wt-wt percent)-based thermosetting molding composition was made according to the method of Example 1 so as to comprise the components shown in Table 2 below:

Table 2

10

Component	Percent by Weight
DEN 431 TM Liquid Epoxy Resin	6.25%
DEN 438 TM Epoxy Resin	6.25%
HRJ-1166 TM Phenolic Novolac	5.35%
Acetone	8.94%
Carnuba Wax	0.54%
Diuron	0.55%
Zinc Stearate	0.36%
Imidazole (2P4MZ)	0.044%
Natural Flake Graphite (Asbury 3562 Flake) [300 micron mesh size; 96.5% purity]	71.72%

Example 4DEN 431TM/438TM/Higher Imidazole-Based
Thermosetting Molding Composition.

- 5 A 2500 gram batch of DEN 431TM/438TM/Imidazole (2P4MZ) [0.065 wt-wt percent]-based thermosetting molding composition was made according to the method of Example 1 so as to comprise the components shown in Table 3 below:

Table 3

10

Component	Percent by Weight
DEN 431 TM Liquid Epoxy Resin	6.25%
DEN 438 TM Epoxy Resin	6.25%
HRJ-1166 TM Phenolic Novolac	5.35%
Acetone	8.92%
Carnuba Wax	0.54%
Diuron	0.55%
Zinc Stearate	0.36%
Imidazole (2P4MZ)	0.065%
Natural Flake Graphite (Asbury 3562 Flake) [300 micron mesh size; 96.5% purity]	71.72%

Example 5DEN 431TM/438TM/No Imidazole-Based
Thermosetting Molding Composition.

- 5 A 2500 gram batch of DEN 431TM/438TM/No Imidazole (2P4MZ)-
based thermosetting molding composition was made according to the method of
Example 1 so as to comprise the components shown in Table 4 below:

Table 4

10

Component	Percent by Weight
DEN 431 TM Liquid Epoxy Resin	6.25%
DEN 438 TM Epoxy Resin	6.25%
HRJ-1166 TM Phenolic Novolac	5.35%
Acetone	8.98%
Carnuba Wax	0.54%
Diuron	0.55%
Zinc Stearate	0.36%
Natural Flake Graphite (Asbury 3562 Flake) [300 micron mesh size; 96.5% purity]	71.72%

Example 6Upper Limit of Conditions selected to preserve the integrity of the filler - Heavy
Batch Load of
Thermosetting Molding Composition

5

A 50 pound batch of EPON 828™/DER 661™-based thermosetting molding composition was made according to the method of Example 1, subject to the following modifications, so as to comprise the components shown in Table 5 below.

The method used was that described in Example 1, except for the following
10 modifications:

1. The mixer (a Hobart Mixer, Model No. V-1401, commercially available from Hobart, Inc., Troy, OH) was set to operate at speed setting number 3 (i.e., about 150 rpm; the same speed setting on different mixers may yield different rpms) during the entire mixing process required to make the composition;
- 15 2. The binder resin system component and the conductive filler component of the thermosetting molding composition were mixed for 30 minutes; and
3. The batch size was 50 pounds (as noted above).

Table 5

20

Component	Percent by Weight
DEN 431™ Epoxy Resin	12.5%
HRJ-1166™ Phenolic Novolac	5.35%
Acetone	8.98%
Carnuba Wax	0.54%
Diuron	0.55%
Zinc Stearate	0.36%
Natural Graphite Flake [80 micron mesh size; 96% purity]	71.72%

Example 7Measuring Flexural Strength of Structure

- 5 Three thin wall structure nominal panels (6 in X 6 in X 0.1 in) were prepared via compression molding techniques, one nominal panel from each of the thermosetting molding compositions made in accordance with Examples 2 (Panel 1), 5 (Panel 2) and 6 (Panel 3). Each of the nominal panels was cut into 3 testing specimens (0.75 in. X 2.25 in X 0.01 in).
- 10 The ultimate flexural strength (e.g., stress on structure immediately prior to cracking) of each of the testing specimens was determined in accordance with the ASTM flexural property procedure published in the Annual Book of ASTM Standards (1997), under the fixed designation D790-97, and under the title "Standard Test Methods for Flexural Properties of Unreinforced and Reinforced Plastics and
- 15 Electrical Insulating Materials."

The results of the ultimate flexural strength for the 3 specimens of each of the panels were averaged to generate an average ultimate flexural strength for the particular panel. The results of this determination of the average ultimate flexural strength for each of the panels are set forth in Table 6 below

Table 6

<u>Panel Number</u>	<u>Average Flexural Strength (psi)</u>
1	3727
2	5130
3	4670

5

Example 8Measuring Electrical Resistivity of Structure

Three thin wall structure thick disks (4 in diameter X 0.1 in thick) were
10 prepared via compression molding techniques, one thick disk from each of the
thermosetting molding compositions made in accordance with Examples 2 (Disk 1), 5
(Disk 2) and 6 (Disk 3).

The electrical resistivity (e.g., reciprocal of electrical conductivity) of
Disks 1 - 3 was determined by applying a known current and measuring a resultant
15 voltage drop, in accordance with the ASTM electrical resistivity measurement
procedure published in the Annual Book of ASTM Standards (1997), under the fixed
designation B193-95, and under the title "Standard Test Methods for Resistivity of
Electrical Conductor Materials."

The results of the electrical resistivity determination are set forth in
20 Table 7 below.

Table 7

<u>Disk Number</u>	<u>Electrical Resistivity (Ohm-cm)</u>
1	0.00095
2	0.0005
3	0.0045

5

Example 9Measuring Thermal Conductivity of Structure

Two thin wall structure nominal plates (6 in X 6 in X 0.1 in) were
10 prepared via compression molding techniques, one nominal plate from each of the
thermosetting molding compositions made in accordance with Examples 5 (Plate 1)
and 3 (Plate 2). Each of the plates was machined into a test specimen (0.5 in. X 0.5 in
X 0.080 in).

The thermal conductivity of a testing specimen from each of Plates 1
15 and 2 was determined over varying temperatures in accordance with the flash method
published in the Annual Book of ASTM Standards (1997), under the fixed designation
E1461-92, and under the title "Standard Test Method for Thermal Diffusivity of
Solids by the Flash Method."

The results of the thermal conductivity determination are set forth in
20 Table 8 below.

Table 8**Thermal Conductivity Calculations**

Sample (No.)	Temp. (C)	Density (gm cm ⁻³)	Specific Heat (W-a-gm ⁻¹ K ⁻¹)	Diffusivity (cm ² sec ⁻¹)	Conduct. (W-cm ⁻¹ K ⁻¹)	Conduct. (BTU *)
Plate 1	23.0	1.546	0.7250	0.04050	0.04538	31.49
	40.0	1.546	0.8150	0.03720	0.04686	32.51
	60.0	1.546	0.9110	0.03400	0.04787	33.22
	80.0	1.546	1.0010	0.03160	0.04889	33.92
	100.0	1.546	1.0820	0.02900	0.04850	33.65
	120.0	1.546	1.1550	0.02640	0.04713	32.70
Plate 2	23.0	1.670	0.7620	0.04820	0.06132	42.54
	40.0	1.670	0.8490	0.04400	0.06237	43.27
	60.0	1.670	0.9430	0.04050	0.06376	44.24
	80.0	1.670	1.0340	0.03740	0.06456	44.79
	100.0	1.670	1.1150	0.03460	0.06441	44.69
	120.0	1.670	1.1820	0.03130	0.06177	42.85

5

* (BTU in hr⁻¹ ft⁻² F⁻¹)

What is claimed is:

1. A thermosetting molding composition comprising:
about 10 to about 30 wt-wt percent of a low viscosity thermosetting
binder resin system, and
about 70 to about 90 wt-wt percent of a filler.
- 5 2. A thermosetting molding composition according to claim 1, wherein
the filler is electrically and thermally conductive.
3. A thermosetting molding composition according to claim 2, wherein
10 the filler is natural graphite flake.
4. A thermosetting molding composition according to claim 2, wherein
the filler is electrically and thermally conductive synthetic graphite.
- 15 5. A thermosetting molding composition according to claim 2, wherein
the low viscosity thermosetting binder resin system comprises an epoxy vinyl ester
resin system.
6. A thermosetting molding composition according to claim 1, wherein
20 the low viscosity thermosetting binder resin system comprises a low molecular weight
phenolic resin and, optionally, an amine curing agent.
7. A thermosetting molding composition according to claim 1, wherein
the low viscosity thermosetting binder resin system comprises a combination of a low
25 viscosity epoxy resin and a low molecular weight phenolic resin curing agent having a
stoichiometric ratio of the phenolic resin curing agent to the epoxy resin in the range
from about 60% to about 110% in the combination.

8. A thermosetting molding composition according to claim 7, wherein the low viscosity epoxy resin is selected from the group consisting of solid or liquid forms of phenolic novolac-based resins, cresolic novolac-based resins, epoxidized phenolic novolac-based resins, epoxidized cresolic novolac-based resins, bi-phenyl-
5 based epoxy resins, dicyclopentadiene-based epoxy resins, bis-F type epoxy resins, bis-A type epoxy resins, and suitable combinations thereof.

9. A thermosetting molding composition according to claim 7, wherein the viscosity of the low viscosity epoxy resin is in the range of about 1400 centipoise
10 to about 50000 centipoise at about 52°C.

10. A thermosetting molding composition according to claim 7, wherein the low viscosity epoxy resin has an epoxy equivalent weight (EEW) in the range of about 165 to about 250.

15

11. A thermosetting molding composition according to claim 7, wherein the low molecular weight phenolic resin curing agent has a softening point in the range of about 60°C to about 90°C.

20 12. A thermosetting molding composition according to claim 7, wherein the low molecular weight phenolic resin curing agent has a molecular weight of less than about 600.

13. A thermosetting molding composition according to claim 7, wherein
25 the low molecular weight phenolic resin curing agent has a hydroxy equivalent weight (HEW) in the range from about 95 to about 115.

14. A thermosetting molding composition according to claim 7, wherein the low viscosity epoxy resin is a mixture of bis-F type epoxy resins, each having an epoxy equivalent weight (EEW) in the range from about 172 to about 181, the low molecular weight phenolic resin curing agent has a hydroxy equivalent weight (HEW)
5 in the range from about 103 to about 107, and the stoichiometric ratio of the phenolic resin curing agent to the epoxy resin is about 70%.
15. A thermosetting molding composition according to claim 7, further comprising about 0.001 to about 3 wt-wt percent of a suitable catalyst system.
10
16. A thermosetting molding composition according to claim 7, further comprising about 0.001 to about 3 wt-wt percent of a suitable catalyst system, wherein the suitable catalyst system comprises about 0.001 to about 1.5 wt-wt percent of urea-based catalyst and about 0.001 to about 0.5 wt-wt percent of an imidazole catalyst.
15
17. A thermosetting molding composition according to claim 16, wherein the urea-based catalyst is diuron and the imidazole-based catalyst is 2-phenyl-4-methyl imidazole.
- 20 18. A thermosetting molding composition according to claim 16, wherein the thermosetting molding composition substantially cures in no more than about 60 seconds at suitable curing conditions.
19. A thermosetting molding composition according to claim 1, further
25 comprising about 0.001 to about 2 wt-wt percent of an internal release agent.
20. A thermosetting molding composition according to claim 1, further comprising about 0.001 to about 15 wt-wt percent of a binder resin system solvent.

21. A method of making a molding composition comprising:
mixing a low viscosity thermosetting binder resin system and a filler
under conditions selected to preserve the integrity of the filler and under conditions
sufficient to wet-out the conductive filler with the binder resin system.
- 5
22. An electrically and thermally conductive structure made from a
thermosetting molding composition according to claim 2.
23. An electrically and thermally conductive structure according to claim
10 22, wherein the structure comprises at least one thin wall section.
24. A structure according to claim 23, wherein the thin wall section has a
thickness in the range from about 0.012 inches to about 0.200 inches.
- 15 25. A structure according to claim 23, wherein the thin wall section has a
strength sufficient to avoid cracking when under a flexural stress of no more than
about 2500 psi.
26. A structure according to claim 23, wherein the thin wall section has an
20 electrical resistivity in the range from about 0.0001 Ohm cm to about 0.01 Ohm cm.
27. A structure according to claim 23, wherein the thin wall section has a
thermal conductivity of no less than about 3 (W/m) K.
- 25 28. A method of making a structure from a thermosetting molding
composition, the method comprising
molding into a desired shape a thermosetting molding composition
according to claim 1, and
subjecting the desired shape to suitable curing conditions to form a
30 structure.

29. A method according to claim 28, wherein the desired shape is substantially cured in no more than about 60 seconds at the suitable curing conditions.

INTERNATIONAL SEARCH REPORT

Intern. al Application No
PCT/US 98/21748

A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C08K3/04 C08K11/00 C08K5/00 H01M4/00 C08G59/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C08K C08G

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	<p>DATABASE WPI Section Ch, Week 8329 Derwent Publications Ltd., London, GB; Class A21, AN 83-714247 XP002094044 & JP 58 098378 A (NIPPON LIGHT METAL CO) , 11 June 1983 see abstract</p>	1-4, 6, 21-28
X	<p>DATABASE WPI Section Ch, Week 9149 Derwent Publications Ltd., London, GB; Class A21, AN 91-357927 XP002094045 & JP 03 239752 A (NITTO DENKO CORP) , 25 October 1991 see abstract</p>	1, 28, 29

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents :

- *A* document defining the general state of the art which is not considered to be of particular relevance
- *E* earlier document but published on or after the international filing date
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INTERNATIONAL SEARCH REPORT

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